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THE CERAMIC MINED PROTONIC/ELECTRONIC CONDUCTING MEMBRANES FOR HYDROGEN SEPARATION

REMARKS

Applicant has carefully reviewed and considered the Office Action mailed on June 27, 2006, and the references cited therewith.

Claims 1-15, 20, and 27-29 are canceled, claims 16-19 and 21-26 are withdrawn, and new claims 30-72 are added.

Restriction to one of the following claims was required: 11, 16, 21 and 24.

As provisionally elected by Applicants representative, David Fonda, on June 14, 2006, Applicant elects to prosecute the invention of Group I, claims 1-15, 20, and 27-29.

The claims of the non-elected invention, claims 16-19, 21-23, and 24-26, are hereby withdrawn. However, Applicant reserves the right to later file continuations or divisions having claims directed to the non-elected inventions.

§112 Rejection of the Claims

Claims 2, 4-7, and 11-15 were rejected under 35 USC §112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter that Applicant regards as the invention. Claim 2, 4-7 and 11-15 have been cancelled.

§102 Rejection of the Claims

Claims 1-15, 20 and 27-29 were rejected under 35 USC §102(b) as being anticipated by Kuenstler et al (Physical Chemical Invstigations), Balachandran et al (Solid State Ionics), and/or U.S Patent No. 5,670,270 to Sten A. Wallin (hereinafter "Kuenstler," "Balachandran," and "Wallin" respectively). Applicant respectfully disagrees that Kuenstler, Balachandran, or Wallin anticipate Applicant's claimed invention as amended.

For a reference to anticipate a claim under 35 U.S.C. §102(b), "each and every element as set forth in the claim [must be] found, either expressly or inherently described, in a single prior art reference." *Verdegaal Bros. v. Union Oil Co. of California*, 814 F.2d 628, 631 (Fed. Cir. 1987), cited in MPEP §2131. The cited references do not teach each and every limitation of Applicant's independent claims, either alone or in combination with each other.

Background

The Kuenstler and Balachandran references discuss electrolytes or material that could be suitable for fuel cell applications. The purpose of fuel cells is to create electricity by capturing electrons that are freed as protons (usually from a gas source) pass through the electrolyte. The electrons that are left behind on one side of the electrolyte are collected. The system doesn't function properly if the electrons also pass through the electrolyte. This leakage is undesirable in fuel cell applications because it makes the fuel cell inefficient.

In contrast, Applicant desires electrons to pass through the material used as an electrolyte. Applicant's application is gas separation which involves moving gas in form of protons through an electrolyte to purify the gas. A source of less pure gas gives up only desired gas protons through the material and leaves undesired ions or impurities behind. The faster the exchange of protons through the material, the more efficient the gas separator. The transfer of protons is dependent in part upon the movement of electrons through the material in the opposite direction. Thus, materials that conduct both protons and electrons at substantially equal rates are desirable.

The Kuenstler reference

Kuenstler investigates BaCeO₃ (barium cerates) with varying doping agents and measures ion conductivity. Kuenstler prepared the doped BaCeO3 material using the mixed oxide method. namely mixing of the powders of oxides and carbonates of the cations followed by calcination at 1200°C for five hours. The calcined powders were pressed into pellets and rods and sintered at 1700°C for five hours in air. In both the calcined powder and sintered samples the x-ray diffraction method showed that the resulting structure was BaCeO₃ with a small amount of CeO₂. Kuenstler suggests that at the high temperature processing of the material, BaO from the BaCeO₃ material evaporates leaving small amounts of CeO2 according to the following formula:

Kuenstler is quick to note that its only small amounts of CeO2 that are formed, and that BaCeO3 is a decent electrolyte for fuel cell applications. The evaporation of BaO leads to the formation of CeO_2 and a deficiency in Ba content of the sample. The Ba deficiency was confirmed by chemical analysis.

Applicant's intended use for its material is as a solid electrolyte with high electrical conductivity and almost pure ionic conductivity. The consequence of BaO evaporation is a reduction in the electrical conductivity. Furthermore, CeO2 that appears as a by product will appear on the surface of the material where the reaction with gas occurs. Thus, is not dispersed as a second phase that has at least one contiguous path through the material.

Kuenstler also discloses that the sintered material is stable in oxidizing and reducing atmospheres. While such a property may be necessary for use in some applications, it is limited in gas separation applications in that it does not possess chemical stability in the presence of CO2 and H₂O, both of which are present in common fuels used in conjunction with a solid electrolyte gas separation applications. The presence of a small and uncontrolled amount of CeO2 in the materials does not provide protection against chemical reaction with CO2 and H2O in the fuel gas which is desirable in Applicant's applications. The presence of CO2 and H2O in both oxidizing and reducing atmospheres causes the BaCeO3 to react with those gases according to the following formulas:

$$BaCeO_3 + CO_2 \implies BaCO_3 + CeO_2$$

$$BaCeO_3 + H_2O \implies Ba(OH)_2 + CeO_2$$

In contrast to Kuenstler, Applicant's invention teaches that the addition of CeO2 to the BaCeO3 improves the stability of the BaCeO3 in the presence CO2 and H2O. As each mole of BaCeO3 reacts to produce one mole of CeO3, an adequate quantity of CeO2 needs to be present in the mixture to provide the stability in the presence of CO₂ and H₂O. While inadequate processing of the raw materials to produce BaCeO3 may result in trace amounts of CeO2, such mixture will be unstable in CO2 and H2O containing gas atmospheres. The subject invention discloses that addition of sufficient quantity of CeO2 to the BaCeO3 provides the required stability.

Furthermore, Applicant's invention teaches a mixture of two ceramic materials, one a proton conducting BaCeO3 and the second an electron conducting CeO2, with each forming a THE CERAMIC MIXED PROTONICAL ECTRONIC CONDUCTING MEMBRANES FOR BY DROGEN SEPARATION

contiguous phase in the sintered structure for use as hydrogen separation membrane. In order to form a contiguous structure, each phase should have a sufficient volume fraction, generally considered to be 30%, for providing independent proton conducting and electron conducting pathways through the membrane for transport of hydrogen, in the form of protons in BaCeO₃ phase accompanied by electronic conduction through the CeO2 phase to maintain electrical charge balance. Thus, a membrane device made of such mixture will function as a hydrogen separation membrane by virtue of differences in the electrochemical potential, i.e. concentration of hydrogen across the membrane without requiring external electric input. Thus the trace amount of CeO₂ present in samples such as those made by Kuenstler will not provide the required stability in CO2 and H2O as well as not function as hydrogen separation membrane.

Independent claim 30 includes the limitation that the electron conducting phase includes at least a portion that is not the product of a reaction involving the proton conducting ceramic phase. The presence of CeO₂ in the Kuenstler reference is solely as the product of the proton conducting ceramic phase (BaCeO₃) reacting during the process of creating the BaCeO₃. Applicant adds additional amounts to achieve its purposes. Knenstler would reduce the amount of CeO₂ present; Applicant desires at least a minimum amount. Kuenstler does not teach adding CeO₂ to the material and thus does not teach that the electron conducting phase includes at least a portion that is not the product of a reaction involving the proton conducting ceramic phase. Accordingly, Kuenstler does not anticipate claim 30.

Independent claim 52 includes the limitation that the electron conducting ceramic phase forming at least one contiguous path between opposing ends of the material such that electrons may pass through the material. Kuenstler does not teach this anywhere in the reference. In fact, as mentioned above, Kuenstler desires a material that does not conduct electrons through the material and small amounts of CeO2 that occur only as a byproduct are insufficient to form at least one contiguous path through the material. Accordingly, Kuenstler does not anticipate claim 52.

Independent claim 72 includes the limitation that the amount of the electron conducting ceramic phase is sufficient to prevent the formation of carbonate in the presence of carbon dioxide and the formation of hydroxide in the presence of water. As stated above, the material disclosed in the Kuenstler reference is reactive to water and carbon dioxide forming hydroxide and carbonate respectively. Small amounts of CeO₂ would not prevent this. Thus, Kuenstler does not teach this limitation and does not anticipate claim 72.

The Balachandran reference

Balachandran discusses mixed-conducting oxides used in applications such as fuel cells, gas-separation membranes, sensors and electrocatalysis. Balachandran teaches a proton conducting membrane Ba(Ce,Y)O₃. This composition exhibits high proton conductivity and high protonic transference number. High transference number inherently implies that it does not possess sufficient mixed conductivity for hydrogen separation. Protonic transference is a ratio of protonic conductivity over (protonic plus electronic) conductivity. High protonic transference equates to low electronic conductivity, or small amounts of electron conducting phase.

The membrane taught by Balachandran is a *single* phase material. While exposure to H₂O and CO₂ may convert some of the BaCeO₃ into CeO₂, it will not form a dual phase material that Applicant claims and will not be capable of mixed conduction for three reasons: 1) the CeO₂ formed by the reaction will not result in a contiguous phase in the structure that is required in a two-phase composite mixed conductor; 2) the CeO₂ resulting from the reaction will be <u>at the expense of BaCeO₃</u> (see earlier reaction equations) and thus destroy the proton conducting phase of the membrane; and 3) the products of the reaction will also contain BaCO₃ or Ba(OH)₂, both of which do not possess either proton or electron conduction and thus degrade the electrical properties in addition to mechanically weakening the membrane structure. Thus, merely exposing BaCeO₃ disclosed by Balachandran to H₂O or CO₂ will not produce a dual phase mixed conductor as taught by the Applicant's invention.

Independent claim 30 includes a multi-phase limitation where the electron conducting phase includes at least a portion that is not the product of a reaction involving the proton conducting ceramic phase. The presence of CeO₂ in the Balachandran reference is part of a *single phase* material. CeO₂ may be present, but solely as the product of the proton conducting ceramic phase (BaCeO₃) reacting during the process of creating the BaCeO₃. It is not present as a *second phase*. Applicant adds CeO₂ to its material as a second phase. The electron conducting phase is present as a purposeful addition to whatever may be there naturally. The natural occurrence of material that is exists only at the expense of another material is not a second phase. Balachandran does not teach the addition of electron conducting phase material in

anticipate claim 30.

addition to what might be present as a by product of a reaction. Thus, Balachandran does not teach that the electron conducting phase includes at least a portion that is not the product of a reaction involving the proton conducting ceramic phase. Accordingly, Balachandran does not

Independent claim 52 includes the limitation that the electron conducting ceramic phase forming at least one contiguous path between opposing ends of the material such that electrons may pass through the material. Balachandran does not teach this anywhere in the reference. In fact, as mentioned above, Balachandran desires a material that does not conduct electrons through the material and small amounts of CeO₂ that occur only as a byproduct are insufficient to form at least one contiguous path through the material. Accordingly, Balachandran does not anticipate claim 52.

Independent claim 72 includes the limitation that the amount of the electron conducting ceramic phase is sufficient to prevent the formation of carbonate in the presence of carbon dioxide and the formation of hydroxide in the presence of water. As stated above, the material disclosed in the Kuenstler reference would be reactive to water and carbon dioxide forming hydroxide and carbonate respectively. Small amounts of CeO₂ would not prevent this. Thus, Balachandran does not teach this limitation and does not anticipate claim 72.

The Wallin Reference

Independent claims 30, 52, and 72 include the limitation that the material is gas impermeable when sintered. Wallin discloses a composite oxygen electrode/electrolyte structure for a solid state electrochemical device having a porous composite electrode in contact with a dense electrolyte membrane. Wallin also teaches the use of doped zirconia and Sr doped BaCeO₃ as suitable ionically conductive material. Wallin specifically teaches preparation of oxygen electrode where the 'term oxygen electrode as used herein refers to the electrode at which oxygen is either reduced or oxygen anions are oxidized' (column 2, lines 51 – 53). The suggested use of BaCeO₃ as the ionically conducting component of the electrode inherently uses the small oxygen ion conduction present in the material. Thus, the disclosed material is suitable for use as an electrode in high oxygen containing atmosphere. While 'the mixture comprises a continuous phase of the ionically-conductive material and a continuous phase of the electronically-conductive material, which form interpenetrating networks with respect to each other' (column 2.

lines 60 - 64), the 'oxygen electrode portion of the structure of the invention comprises a *porous* solid-solid mixture' (column 2, lines 55 - 58) of the two phases.

Applicant's invention discloses a proton conducting dense membrane comprising an interpenetrating network of a proton conducting phase, BaCeO₃ and an electron conducting phase, CeO₂ to transport hydrogen ion (proton) in highly reducing gas atmosphere by ionic conduction. A porous structure as disclosed by Wallin will allow molecular diffusion of all gas species from one side to the other and thus prevent separation of high purity hydrogen as taught by Applicant's invention. Accordingly, Wallin does not teach a gas impermeable material. Applicant respectfully requests that Examiner's 102 rejection be withdrawn.

Conclusion

The cited references teach *single phase* materials that are reactive in the presence of water and carbon dioxide. Applicant claims a multi-phase material where the second or electron conducting phase prevents the reaction of the first phase material in the presence of water and carbon dioxide. This second or electron conducting phase allows for electrons to pass through the material. The cited references do not teach this. Applicant's material is also substantially gas impermeable when sintered. The cited references do not teach this novel combination. Accordingly, Applicant respectfully submits that claims 30-72 are in condition for allowance and notification to that effect is earnestly requested. The Examiner is invited to telephone Applicant's attorney (801-978-2186) to facilitate prosecution of this application.

If necessary, please charge any additional fees or credit overpayment to Deposit Account No. 50-358650-3586

Respectfully submitted,

S. ELANGOVAN

By his Representatives,

801-978-2186

Date 10/27/2006

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